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A DROP OF SPECTROSCOPY*

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Summary of the work, awarded with the prize of the 2nd best poster presentation by D. Terwagne at the General Scientific Meeting of the BPS at the Université Libre de Bruxelles on May 21, 2008

Abstract

When a low viscosity oil droplet is laid onto the surface of a high viscosity oil liquid, it stays at rest for a moment before coalescence. The coalescence can be delayed and sometimes inhibited by injecting fresh air under the droplet. This can happen when the surface of the bath oscillates vertically. In this case the droplet basically bounces on the interface. We observe that the conditions for bouncing depends on the frequency, more precisely we observe resonance when the eigenfrequency of the droplet is excited. In some conditions, a droplet presents a non axi-symmetric mode of deformation. That leads to a rotation of the droplet and to a horizontal displacement.

Introduction

The manipulation of tiny amounts of liquid plays a key role in any micro-device used in microfluidic applications. A droplet is often laid onto a substrate where it interacts with its environment. Some liquid may be lost onto the substrate or the separation with the surrounding liquid may be difficult. The idea is to manipulate the droplet without touching it [1] or in other words, to transport the droplet using another fluid, for example air. Couder *et al* discovered that it is possible to make an oil droplet bouncing onto a vibrating bath of oil [2,3,4]. Some air is squeezed between the droplet and the bath avoiding coalescence.

The experiment

Low viscosity (1.5, 10 and 100 cSt) silicon oil droplets are placed on a high viscosity (1000 cSt) oil bath that vibrates vertically (Fig. 1). The viscosity difference ensures that the droplet is more deformed than the bath interface. The bath oscillates according to a sinusoidal motion $A\sin(2\pi ft)$ with an amplitude A and a frequency f between 20 Hz to 400 Hz.



Fig. 1: Experimental setup is composed of an oscillating plate on which a container is filled with silicon oil of viscosity 1000 cSt. Silicon oil droplets of viscosity of 1.5, 10 and 50 cSt are laid onto the bath.

The bouncing of the droplet only occurs when the maximal acceleration of the bath oscillation $\gamma_m = A\omega^2$ is higher than a threshold value γ_{th} . This threshold depends among others on the forcing frequency *f*, the droplet radius *R*, and the viscosity *v*. For very small droplets ($R < \kappa^{-1} = \sqrt{\gamma / \rho g}$ (capillary length)), γ_{th} is close to 1*g*, that corresponds to the lift-off threshold for an inelastic solid sphere deposited on a vibrating solid plate. In Fig. 2, the amplitude threshold $A_{th} = \gamma_{th} / \omega^2$ is represented as a function of the forcing frequency *f* for droplets of viscosity of 1.5 cSt and R = 0.765, 0.812 and 0.931 mm. As shown in this figure, a scaling has been found for the amplitude threshold A_{th} and the frequency *f*, which will be explained later.



Fig. 2: Dimensionless amplitude threshold $A_{th} f_c^2 / g$ as a function of the reduced forcing frequency f / f_c , where A_{th}^* is the amplitude of the bath vertical motion corresponding to the reduced acceleration Γ_{th} and $f_c = \sqrt{\sigma/M}$ the capillary frequency of a droplet of mass $M = 4/3\pi\rho R^3$. Bullet, square and cross symbols correspond to droplet radius R=0.765 mm, 0.812 mm and 0.931 mm respectively. The droplet viscosity is v=1.5 cSt. Specific droplet modes of deformation $\ell = 2$, $\ell = 3$ and $\ell = 4$ with m = 0 corresponding to spherical harmonics Y_l^m are observed in the minima zones. Boundaries between those zones correspond to maxima in the amplitude threshold curve.

Resonant modes

The dimensionless amplitude curves $A_{th}f_c^2/g$ as a function of the reduced forcing frequency f/f_c present regularly spaced minima that, in some way, correspond to a resonance of the system: a minimal energy supply is required to sustain the periodic bouncing motion. Those minima tend to disappear when the viscosity of the droplet is increased. Moreover, specific modes of deformation are observed at minima of the $A_{th}f_c^2/g$ curve. They are analogous to the natural modes of deformation expressed by Rayleigh in terms of spherical harmonics Y_l^m . Modes m = 0 and $\ell = 2, 3, 4$ and 5 are observed (Fig. 3).



Fig. 3: Various deformation modes of a bouncing droplet (v=1.5 cSt, *R*=0.765 mm) observed with a high-speed camera for various forcing frequencies. The first line (resp. 2nd, 3rd, 4th) displays a mode Y_2^0 (resp. Y_3^0 , Y_4^0 , Y_5^0) with m = 0 (axisymmetry). The forcing frequency is 50 Hz (resp. 160 Hz, 275 Hz, 300 Hz) and the reduced acceleration $\Gamma = 0.3$ (resp. $\Gamma = 2$, $\Gamma = 6$, $\Gamma = 21$). First and second columns represent snapshots at two different times of the oscillation. The spherical harmonic solution (on the right of each picture) is superposed to the experimental pictures (on the left of each picture).

Indeed, the bouncing droplet may be considered as an oscillating system analogous to the damped driven harmonic oscillator: surface tension is the restoring force and viscosity is the damping process. The dimensionless ratio between both is the Ohnesorge number $Oh = v\sqrt{\rho/\sigma R}$, which is $\ll 1$ in our case. When $Oh \ll 1$, the viscous damping is negligible and resonance is important. Damping increases as Oh gets closer to 1 and minima tend to disappear.

Since those oscillations are due to surface tension, natural frequencies scale as the capillary frequency $f_c = \sqrt{\sigma/M}$ where $M = 4/3\pi\rho R^3$. Moreover, at the bouncing threshold, the vertical force resulting from the droplet deformation exactly balances the gravity. One may define a characteristic length $L = g/f_c^2$ corresponding to the free fall distance during the capillary time $1/f_c$. As shown in Fig. 2, the threshold amplitude A_{th} scales as the length L, whatever the droplet size.

A model of the bouncing droplet has been developed for the first mode of deformation Y_2^0 in [5]. The model takes into account the effect of the droplet deformation on the drainage of the thin air layer in between the droplet and the bath. The first minimum is explained by a resonance of the damped driven harmonic oscillator that the droplet constitutes when the drainage of the film is considered. We show that this minimum disappears for higher viscosities. Then a cut-off frequency has been deduced above which this mode of deformation cannot be used to bounce, it happens at the first maximum. This frequency corresponds to the resonant frequency of the droplet without considering the drainage of the film.

Moreover, the dispersion relation prescribes the natural "Rayleigh" frequency f_R related to a ℓ -mode :

$$\left(\frac{f_{R}(\ell)}{f_{c}}\right) = \frac{1}{3\pi}\ell(\ell-1)(\ell+2)$$

Usually $f_{\text{Resonnance}} = \alpha f_R(\ell)$; α being a multiplication factor which depends on the geometry of the excitation [6, 7]. In this case, α is find equal to 1.15 and those frequencies correspond to the different modes boundaries (maxima in Fig.2). In Fig. 2, arrows indicate the Rayleigh frequencies, which cannot be directly related to inflections in the threshold curve.

When $f = \alpha f_R(\ell)$, the droplet stores the whole energy provided by the oscillating bath in deformation and dissipates it due to enhance internal motion : the droplet resonates. It

is impossible to make the droplet bouncing in the mode $(\ell, m = 0)$, A_{th} should diverge. Experimentally, a maximum is found at $f = \alpha f_{R}(\ell)$ as $m \neq 0$ modes are excited.

Displacement mode : The Roller

At the first maximum of the $A_{lh}^{*}(f/f_c)$ curve $(f/f_c=1)$, the droplet moves along a linear trajectory. A movie of the moving droplet has been recorded using a high speed camera (Fig. 4). The images are separated by 1 ms. The deformation is not axi-symmetrical as in the resonant minima of the threshold amplitude curve. On the other hand, the mode is related to $\ell = 2$ and m = 1. Two lines of nodes that are orthogonal characterize this mode. It results the existence of two fixed points located on the equator of the droplet. As the line of nodes does not follow the axi-symmetrical geometry, those fixed points move. More precisely, they turn giving the droplet a straight direction motion. This is a way to observe the Y_2^1 mode of deformation, which is generally degenerated with the Y_2^0 mode. The first maximum corresponds to the resonance of the $Y_{\ell=2}^m$ mode of deformation. Due to the air film dynamic, a droplet in the Y_2^0 mode cannot bounce [5] while a droplet in the Y_2^1 mode can as the droplet is rolling.



Fig. 4: Rolling motion of the droplet. Mode Y_2^1 of a bouncing droplet (v=1.5 cSt, R=0.765 mm) observed at f=115 Hz and Γ = 4.5. Frames are separated by 1 ms. The droplet is rolling towards the left.

The initial horizontal speed v of the 1.5 cSt viscosity droplet in the mode ($\ell = 2, m = 1$) has been measured for various frequencies and amplitudes of the bath. A scaling is found when considering that the phenomenon can occur only above a cut-off frequency f_0 and an amplitude threshold A_{th} as $v = 2\pi\beta(A - A_{th}(f))(f - f_0)$, where β is a constant. The initial speed is represented versus the characteristic speed $v^* = (A - A_{th}(f))(f - f_0)$ in Fig. 5.



Fig. 5: Initial speed of the roller for various frequencies (indicated in the legend) as a function of the characteristic speed v^* .

Conclusion

The deformation of low viscosity bouncing droplets is emphasized on a high viscosity bath: the droplet oscillations are much less damped than the bath oscillations. Depending on the forcing frequency, droplets need a different amount of supplied energy to achieve a sustained periodic motion. When the forcing frequency corresponds to a multiple of the eigenfrequency of the bouncing droplet $(f/f_c \sim \ell - 1)$, the supplied energy is mainly lost into internal motions, so the threshold acceleration A_{th} is maximum. On the other hand, A_{th} is minimum when $f/f_c \sim \ell - 3/2$.

A new self-propelled mode has been discovered for forcing frequencies between the $\ell = 2$ and $\ell = 3$ modes for a sufficiently low droplet viscosity. This excited mode is a non-axisymmetrical mode Y_2^1 characterized by an internal rotation of the fluid inside the droplet. The droplet rolls over the vibrated bath! This droplet displacement technique is more adapted to large low-viscosity droplets; it is therefore of considerable interest for microfluidic applications: manipulating aqueous mixtures without touching them.

Aknowledgements

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